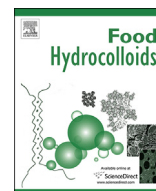


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Food Hydrocolloids

journal homepage: www.elsevier.com/locate/foodhyd

Predicting sensory perceptions of thickened solutions based on rheological analysis

Qi He ^{a, b}, Joanne Hort ^c, Bettina Wolf ^{a, *}^a Division of Food Sciences, The University of Nottingham, Sutton Bonington Campus, Leicestershire, LE12 5RD, UK^b Mondelez International, Reading Scientific Service Limited, Pepper Lane, Reading, RG6 6LA, UK^c International Centre for Brewing Science and Sensory Science Centre, Sutton Bonington Campus, University of Nottingham, Loughborough, Leicestershire, LE12 5RD, UK

ARTICLE INFO

Article history:

Received 25 January 2016

Received in revised form

3 May 2016

Accepted 8 May 2016

Available online 13 May 2016

Keywords:

Mouthfeel

Flavour

Sweetness

Xanthan

Dextran

Thin film rheology

APCI-MS

ABSTRACT

The sensory perceptions of sweetened, flavoured and thickened solutions prepared from xanthan, dextran, sucrose and banana flavour were evaluated and correlated to rheological parameters. The primary aim of this research was to evaluate the relevance of viscosity measured at low shear or at high shear for predicting sensory perceptions. Additionally considered were extensional viscosity estimated from filament thinning experiments and complex shear viscosity. The design of experiments included two groups of 5 samples matched at low shear rate and high shear rate, respectively. Mouthfeel perceptions were well correlated to low shear viscosity, however, including high shear viscosity or extensional viscosity as an additional model parameter improved the predictive quality of the models for thickness, stickiness and mouth coating. Stickiness and mouth coating were better correlated to extensional viscosity than low shear viscosity, although a model including both parameters predicted stickiness and mouth coating best. The complex viscosity at 100 rad/s was also highly correlated to the perception of thickness. Since correlations were not improved over steady shear parameters, complex viscosity was not considered in models based on more than one rheological parameter. Flavour was also scored during sensory evaluation and sweetness and overall flavour were highly correlated. The results of this study have highlighted that there is no single rheological parameter that will ultimately correlate to a range of mouthfeel perceptions. For certain mouth feel perceptions a model comprising shear and extensional rheological parameters will have higher predictive power than a model solely based on shear rheological parameters.

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1. Introduction

It has long been the ambition of the food industry to correlate sensory perception with instrumentally measured parameters in order to simplify and lower the cost of product development activities. Running sensory panels is expensive whereas, once appropriate equipment is accessible, measurement of physical parameters is often seen as an efficient alternative and as a result numerous studies have been performed with the aim of establishing methods that reflect in-mouth sensory properties using physical parameters. In terms of liquid and liquid-like products the study of rheological properties has become a major interest.

Szczesniak and Farkas (1962) published one of the earliest studies on the correlation of the mouthfeel of liquid and semi-liquid foods with their shear rheological properties. Using a wide range of gum solutions of about the same low shear viscosity (around 1.2 Pa s), these authors suggested that within the shear rate range of 0–100 s^{−1} perceived sliminess is negatively correlated to shear thinning rate. However, the data in their research do not provide sufficient insight to know whether it is the rate of shear thinning, the shear rate at which shear thinning starts or the viscosity at shear rates relevant to in-mouth perception that determines sliminess (van Vliet, 2002). It was then Wood (1968) who for the first time studied the flow conditions in the oral evaluation of liquids. In his research, the shear rate chosen for the measurement of viscosity to relate to the perception of thickness was determined by asking subjects to compare the thickness of cream soups to glucose syrups which were of shear thinning and

* Corresponding author.

E-mail address: Bettina.wolf@nottingham.ac.uk (B. Wolf).

Newtonian flow behaviour, respectively. He then postulated that the shear rate at which the viscosity curve of a soup and a syrup with similar perceived thickness crossed is pertinent to thickness perception and this shear rate is 50 s^{-1} (Wood, 1968). Later on, a similar approach was applied to a wider range of food products (Shama, Parkinson & Sherman, 1973; Shama & Sherman, 1973) to investigate whether it is in fact a range of shear rates and shear stresses that is relevant to perception of thickness. It was found that the stimulus associated with oral viscosity perception of liquid and semi-liquid foods embraces a wide range of shear rates from 10 to 1000 s^{-1} strongly depending on the viscosity of the products. Data showed that low viscosity liquids with a measured viscosity of 0.1 Pa s and below are orally evaluated at a constant shear stress of roughly 10 Pa while higher viscosity liquid foods of 10 Pa s and above are assessed at a constant shear rate of approximately 10 s^{-1} (Shama & Sherman, 1973). Later Christensen (1979) suggested that perceived viscosity is represented by an averaged viscosity over a range of shear rates rather than by viscosity measured at a single shear rate. This finding is based on a study using low, medium and high molecular weight carboxymethyl cellulose (CMC) solutions prepared to the same low shear viscosity value. The high molecular weight CMC solution was perceived as thinner than the low or medium molecular weight CMC solutions. The high molecular weight solution was more shear thinning and its viscosity averaged over a range of shear rates in the shear thinning regime lower than for the low or medium molecular weight solutions, thus reflecting the sensory findings. Koliandris et al. (2010) found in a study based on dextran and guar gum solutions that thickness perception was affected by both the degree of shear-thinning and the viscosity value at high shear rate (10^5 s^{-1}).

Viscosity measured in small deformation oscillatory shear has also been related to oral thickness (Bistany & Kokini, 1983; Richardson, Morris, Ross-Murphy, Taylor and Dea, 1989). Richardson, Morris, Ross-Murphy, Taylor, and Dea (1989) found that perceived thickness correlated well to measured (large deformation) shear viscosity for “true solutions”, which they defined as solutions which do not exhibit a yield stress. However, the oral thickness of extremely shear thinning samples such as of weak gels was underestimated. Small deformation measurements of dynamic viscosity under oscillatory shear at a single frequency of approximately 50 rad/s were subsequently found to highly correlate to sensory panel scores for perceived thickness not only for weak gels but also for “true solutions” ($r = 0.95$).

To further the understanding of oral perception, Kokini et al. (1977), Kokini and Cussler (1983) developed a model to calculate oral shear stress exerted onto liquid foods manipulated between the tongue and the upper palate by representing the oral cavity as a parallel plate geometry. The model is based on the assumption that the shear rate dependent viscosity behaviour of a liquid food follows power law behaviour. Elejalde and Kokini (1992) successfully used this model to correlate oral shear stress to perceived thickness when evaluating different power law hydrocolloid solutions and other liquid foods. There is still much debate about magnitude and distribution of shear rate and shear stress in the oral cavity as well as the effect of non-rheological parameters on the perception of sensory texture (Malone, Appelqvist & Norton, 2003).

It has also been suggested that the transient shear rate in mouth can reach values as high as 10^5 s^{-1} (Nicosia & Robbins, 2001). Following this insight researchers have started to consider high shear viscosity in relation to the in-mouth behaviour of foods (Davies & Stokes, 2008; Koliandris et al., 2010; de Vicente, Stokes & Spikes, 2006). Malone, Appelqvist, and Norton (2003) further introduced the idea of the friction behaviour between tongue and palate to be relevant to sensory texture perception. Oral friction may be mimicked through appropriate design of the friction

partners in commercial tribometers (Bongaerts, Fourtouni & Stokes, 2007) or purpose build friction devices (Ranc, Servais, Chauvy, Debaud & Mischler, 2006; Dresselhuys, de Hoog, Stuart, & van Aken, 2008). Whilst most publications dealing with the tribological properties of liquid and semi-liquid foods are void of substantial sensory data and appropriate correlations, it has been suggested that creaminess of emulsion based foods correlates well to friction coefficients (Chen & Stokes, 2011).

In addition to behaviour in shear flow, most often assumed to be the prevailing flow pattern during the oral processing of liquid and semi-liquid foods, some researchers have provided evidence that extensional flow properties could be similarly important (Debruijne, Hendrickx, Alderliesten & Delooff, 1993; van Vliet, 2002; Koliandris et al., 2011). The concept is that foods are initially compressed between the tongue and the palate similar to squeezing flow between two parallel plates. Then, on separation, biaxial extensional flow develops as if the plates were lubricated (Chatraei, Macosko, & Winter 1981). However, the relationship between extensional flow behaviour and sensory perception has barely been investigated. One exception is the use of Boger fluids to study the relationship between the perception of saltiness and extensional viscosity (Koliandris et al., 2011), however, mouthfeel was not considered.

The research presented here is not only concerned with texture perception but also with flavour perception. It is normally recognized that perceived flavour decreases as product thickness increases, for example, due to the addition of hydrocolloid thickeners (Christensen, 1980; Izutsu, Taneya, Kikuchi & Sone, 1981; Baines & Morris, 1987; Cook, Hollowood, Linforth, & Taylor, 2001). The possible reason for this could be that tastant molecules are inhibited from contacting the taste receptors on the tongue, and further prevent the interactions between taste and aroma and therefore flavour perception is less intense (Bayarri, Smith, Hollowood, & Hort, 2007). Baines and Morris (1987) found that perceived intensity of sweetness and strawberry flavour was significantly affected by the addition of guar gum at concentrations above the random coil overlap concentration (c^*) with no effect observed below c^* . Based on an analysis of the possibility of an interaction between flavour molecules and the polymer, and restricted diffusion, the researchers came to the conclusion that the dominant effect is less efficient mixing in solutions with a high degree of polymer entanglement, i.e., above c^* , which inhibits replenishment of surface depletion. Mixing presumably refers to mixing with saliva although the authors have not stated this specifically. Later, Malkki, Heini & Autio (1993) presented data from which they concluded that, depending on the nature of a thickener, chemical bonding or adsorption between flavour and thickener molecules may occur and affect flavour perception. They also mention the possibility that other rheological parameters than shear viscosity have influenced perception. Roberts, Elmore, Langley, and Bakker (1996) reported that it was both the binding of flavour molecules to the polymers and the physical inhibition of volatile movement limiting flavour perception above c^* . Hollowood, Linforth, and Taylor (2002) found that although perceived strawberry and almond flavour as well as sweetness was greatly reduced when hydroxypropyl methylcellulose (HPMC) was added at concentrations above c^* , the concentration of volatile compounds released into the breath (measured using MS Nose™) was not affected by the increased viscosity. The results were explained by reduced availability of free water in solution with increasing HPMC concentration leading to decreased sweetness perception and further affecting the intensity of flavour perception. The main interest of their research was saltiness perception which was inversely related to viscosity measured at low shear rate ($10\text{--}50 \text{ s}^{-1}$).

Surveying the literature has shown that there are still knowledge gaps with regard to which are the most useful rheological parameters relating to oral perception. In an attempt to contribute further knowledge to this field of research we have designed this systematic study concerning the perception of mouthfeel and flavour and its relationship to low and high shear viscosity. We have extended the rheological characterisation of the study samples to include the methods of filament thinning and small amplitude oscillatory shear to assess whether consideration of extensional viscosity and complex viscosity would improve correlations between physical and sensory data solely based on steady shear viscosity data.

2. Materials and methods

In order to have a wide design space in terms of level of low and high shear viscosity, the study samples comprised aqueous solutions of xanthan gum and dextran, or mixtures thereof. Due to its rod-like molecular structure, xanthan gum is extremely shear thinning and considered as forming the most shear thinning gum solutions available (Holme, Hall, Speers, & Tung, 1988). The factor between low shear and high shear viscosity may be several decades. Dextran on the other hand is Newtonian, even at shear rates as high as 10^5 s^{-1} (Koliandris et al., 2010). In the range of concentrations used in this study, mixtures of these two hydrocolloids formed single phase solutions.

In the initial stages of this research the composition of the study samples was devised based on steady shear rheology carried out at 20°C and this same measurement temperature was applied for additional evaluation of the study samples in dynamic oscillatory shear. It was thought convenient not to consider in mouth temperature for the sample design stage of the research because the samples were presented to the sensory panel at room temperature ($20^\circ\text{C} \pm 2^\circ\text{C}$). Therefore the predictive models for sensory perceptions presented are also based on shear rheology data acquired at 20°C . At a later stage it appeared interesting to include filament break up as a method to characterise the behaviour of the study samples in extensional flow. As these tests were not part of the design stage, measurement temperature was chosen as in mouth temperature and therefore extensional viscosity in the predictive models refers to data valid at 37°C . This inconsistency in temperature is unlikely to have an impact on the principle nature and power of the models presented.

2.1. Materials and solution preparation

Food grade xanthan Keltro RD (CP Kelco, UK) and low molecular weight dextran (10 kDa, Meito Sangyo, Japan) were selected for this study and aqueous gum solutions were prepared with bottled water (Evian, Danone, France). The samples were flavoured with sucrose (Tate & Lyle, UK) and banana flavour (isoamyl acetate, Firmenich, Geneva, Switzerland). To determine the gap error in the thin film rheological test, silicon oil (100/200 cSt, Dow Corning, USA) was used as the Newtonian reference following published protocol (Davies & Stokes, 2008). Samples were prepared by mixing stock solution of xanthan, dextran and a mixture of sucrose and isoamyl acetate (IAA) on a rolling bed mixer for 24 h at 4°C . A stock solution of 2% (w/w) xanthan was prepared by dispersing the dry powder into water, heating to 85°C followed by stirring for 1 h at this temperature and further mixing on a rolling bed mixer overnight at 4°C . 38% (w/w) dextran stock solution was prepared by dispersing and dissolving the dry powder into water using a magnetic stirrer for 3 h. Additionally, using water as solvent a stock solution of 30% (w/w) sucrose and 100 ppm IAA was prepared by mixing on a magnetic stirrer. The stock solutions and all samples

were used within one week of preparation.

2.2. Shear rheology

All shear rheological measurements were conducted using a rotational rheometer (MCR301, Anton Paar, Austria) and measurement temperature was 20°C . Steady state shear viscosity data were acquired up to a shear rate of 10^5 s^{-1} with a 50 mm diameter parallel plate geometry at the three gap heights of 500 μm , 50 μm and 30 μm applying the technique of “thin film rheology” (Davies & Stokes, 2008). Flow curves obtained were fitted to a log-log model (Equation (1)) enabling calculation of the apparent viscosity at any shear rate in the measurement range applied.

$$\log(\eta) = \delta + \frac{\alpha}{(1 + e^{(\beta - \gamma \times \log(\dot{\gamma}))})} \quad (1)$$

where η is the apparent shear viscosity (Pa s), 10^δ is the high shear viscosity asymptote (Pa s), $10^{(\alpha + \delta)}$ is the low shear viscosity asymptote (Pa s), β/γ is the point of inflection, and $\dot{\gamma}$ is the shear rate (s^{-1}).

Small deformation oscillatory shear data were acquired with a 50 mm diameter cone-and-plate geometry by initially carrying out an amplitude sweep to explore the linear viscoelastic (LVE) range using a strain sweep from 0.1 to 1000% at the angular frequency of 10 rad/s. Then an angular frequency sweep from 0.1 to 100 rad/s within the LVE was carried out (strain of 1%).

2.3. Extensional rheology

To evaluate the rheological behaviour in a predominantly extensional flow field filament break up tests were conducted using commercial filament breakup equipment (Haake CaBER1 extensional rheometer, Thermo Haake GmbH, Karlsruhe, Germany). These measurements were conducted at a later stage in the research to relate to the in-mouth characteristics of the designed samples and therefore these data were acquired at 37°C . The principle of the method is to record the thinning of the filament at mid-point following stretching between two plates. Using a 1 mL syringe fitted with a needle a droplet of sample was loaded into the gap between the upper and bottom plates of the instrument (6 mm plate diameter, 3 mm initial gap). The sample was then stretched linearly in 50 ms to a 10 mm long filament. The diameter of the filament was measured at mid-point of the final gap height (D_{mid}) from D_0 at time t_0 (when the top plate reaches the final gap) to t_b (when the filament breaks). The results were analysed for steady state extensional viscosity by fitting different models to the filament thinning curves. More detailed analysis of the filament thinning data, not exploited for correlation to sensory perceptions, is provided in the electronic supplementary file. To calculate the apparent extensional viscosity (see Equation (2)) the surface tension of the sample was required, for which the value of water (70 mN/m) at 37°C (Vargaftik, Volkov, & Voljak, 1983) was used for all as some of the samples were of high viscosity rendering it difficult to acquire surface tension data. For each sample, at least 10 replicate measurements were performed and 3 representative sets of data averaged to obtain one set of results.

$$\eta_E = -\frac{\sigma}{dD_{mid}/dt} \quad (2)$$

η_E is extensional viscosity; σ is the surface tension; dD_{mid}/dt is the rate at which the mid-point filament diameter decreases with time.

2.4. Sensory evaluation

A modified Quantitative Descriptive Analysis was selected as the sensory evaluation technique (Stone, 1999). Ten experienced panellists (age range from 45 to 70: 1 male and 9 female) attended altogether three types of sessions: three training sessions, two practice rating sessions and three final rating session. In the training sessions, four out of the ten samples were selected so the panellists could familiarise themselves with the definition of the pre-determined attributes and the methods used to evaluate these attributes adapted from a previous study in which they had participated (Zhang, 2009). Altogether the panellists generated seven attributes which can be divided into two categories: Mouthfeel and flavour & taste. Their definitions including protocols of evaluation on a 'Not at all' to 'very' 10 point scale are summarised in Table 1. Two practice rating sessions were carried out to monitor the performance of the panellists in terms of scale use and consistency of attribute rating: no further training was deemed necessary. In the final rating sessions, all ten samples were presented monadically in triplicate according to a randomised partially balanced design and panellists were asked to rate of all the attributes on continuous lines scales labelled low to high. A 2 min break was given between each sample to avoid effects of sensory fatigue and carry over. All of the samples were coded with random three digit numbers and the data were collected using appropriate software (Fizz, Biosystems, France) in sensory booths at the University of Nottingham Sensory Science Centre. Panellists were instructed to use plain crackers (99% Fat Free, Rakusen's UK) and still mineral water as palate cleansers before each sample. All sensory tests were carried out at room temperature (20 °C).

2.5. Flavour release

Real time in-nose release of IAA during consumption of the samples was measured using Atmospheric Pressure Chemical Ionisation-Mass Spectrometry (APCI-MS) (Micromass, Manchester, UK). Three panellists were asked to place 10 mL of sample into their mouth with a spoon and chew and rubbing the samples with tongue with the mouth closed while breathing normally into the APCI-MS nasal sampling tube. A training session before the measurement was used to make sure the panellists were consuming the samples in a consistent manner. Air from the nose was sampled at 30 mL/min and the release of IAA was followed by monitoring *m/z* 131 (the mass to charge ratio for the molecular ion). Breath by breath data were recorded as peak heights and the data were then analysed to generate two parameters, the maximum volatile release (Imax) and the cumulative area under the 1.5 min release

profile (AUC). Measurements were conducted on all samples in duplicate and palate cleansers as well as breaks were the same as described for sensory evaluation. All flavour release measurements were carried out at room temperature (20 °C).

2.6. Statistical analysis

Statistical analysis of the data was performed using SPSS (version 16, IBM, USA). Analysis of variance (ANOVA) was applied to explore if there were any differences between the samples in terms of sensory properties. Post hoc and, where appropriate, a Tukey's HSD test was performed to find out which samples were significantly different to the others ($\alpha = 0.05$). In addition, Pearson correlation coefficients (*r*) were calculated between perceived sensory attributes and measured viscosity. Experimental design software used to generate the samples was also employed to build regression models that express sensory scores in significant terms relating to the different physical properties. In model building, several parameters were used to examine the quality of models such as: the predicted R^2 was used indicates how precise the model is at predicting the results from the samples tested. The adjusted R^2 is used to indicate how well the model would describe variation outside the sample range. The adequate precision is used to indicate signal to noise ratio.

3. Study sample design and rheological properties

3.1. Sample design

The objective of the sample design was to obtain two groups of samples based on shear viscosity: In the first group (Group 1), all of the samples were designed to have the same viscosity at low shear rate (50 s^{-1}) but different viscosity at high shear rate (10^5 s^{-1}); in the second group (Group 2), all of the samples were designed to have the same high shear viscosity (10^5 s^{-1}) but different low shear viscosity (50 s^{-1}). To achieve this, experimental design software (Design-Expert version 7.1.Stat-Ease, USA) was used to evaluate the relationship between polymer concentration (for xanthan and dextran) and the viscosity at both low and high shear rate based on a D-optimal Response Surface design. Within the experimental design, the concentration of xanthan and dextran was varied from 0 to 1%w/w and 0 to 30%w/w, respectively. 16 samples altogether were generated by the software, with duplicates at each corner point and at the centre point, see Table 2. The low and high shear viscosity values of these samples were measured and fed into the software to generate predictive models for shear viscosity based on Equation (1). The model parameters are included in Table 2 and as

Table 1
Attributes including their definitions and protocol as defined by the panel. A 10 point continuous line scale was used for all attributes.

| | Attribute | Definition | Protocol |
|-----------------|---------------------|--|--|
| Mouthfeel | Initial thickness | The pressure needed to press the sample between the tongue and the palate. | Put a spoonful of sample onto the tongue, gently press the tongue against the palate 3 times. |
| | Thickness in mouth | The pressure taken to move the sample between the tongue and the palate. | Put a spoonful of sample onto the tongue, move the sample in the mouth, rub the tongue for 5 times. |
| | Stickiness on lips | The pressure to separate the sample from the lips. | Use lips to take a tip of sample (avoid touching from lips), and hold there for 5 s, then separate the lips for 3 times. |
| | Stickiness in mouth | The elasticity between the tongue and the palate. | Put a spoonful of sample onto the tongue, gently press the tongue against palate and hold there for 3 s and then separate for 5 times. |
| | Mouth coating | The amount of residues left in the oral cavity after swallowing. | Put a spoonful of sample into the mouth, move around the tongue and chew the sample for 5 times and swallow. |
| Flavour & taste | Overall flavour | The overall intensity of flavour perceived. | Put a spoonful of sample into the mouth, move around the tongue and chew the sample for 5 times and swallow. |
| | Overall sweetness | Overall intensity of sweetness of the samples. | Put a spoonful of sample into the mouth, move around the tongue and chew the sample for 5 times and swallow. |

Table 2

Composition of the 16 samples used in this study and log-log model (Equation (1)) parameter values. The correlation coefficient R^2 equals 0.99 for all 16 fits. The final two columns show shear viscosity at 50 s^{-1} (η_L) and 10^5 s^{-1} (η_H) based on the log-log model fit.

| Sample no. | Concentration xanthan dextran | | α | β | δ | γ | η_L | η_H |
|------------|-------------------------------|---------|----------|---------|----------|----------|----------|----------|
| | % (w/w) | % (w/w) | | | | | Pa s | Pa s |
| 1 | 0.20 | 30.00 | 2.861 | −0.126 | −1.288 | −0.816 | 0.221 | 0.058 |
| 2 | 1.00 | 30.00 | −5.862 | −0.233 | 4.752 | 0.634 | 1.363 | 0.120 |
| 3 | 1.00 | 0.00 | −7.965 | 0.164 | 4.950 | 0.468 | 0.562 | 0.006 |
| 4 | 0.20 | 30.00 | −3.366 | 0.023 | 1.974 | 0.724 | 0.241 | 0.050 |
| 5 | 0.20 | 15.00 | −3.533 | 0.525 | 1.316 | 0.701 | 0.096 | 0.009 |
| 6 | 0.60 | 30.00 | −5.365 | −0.196 | 4.404 | 0.639 | 0.693 | 0.071 |
| 7 | 0.20 | 0.00 | −4.104 | 0.721 | 1.069 | 0.611 | 0.050 | 0.002 |
| 8 | 0.60 | 0.00 | −6.584 | 0.349 | 3.570 | 0.514 | 0.271 | 0.004 |
| 9 | 0.60 | 15.00 | −6.077 | 0.085 | 3.887 | 0.573 | 0.382 | 0.015 |
| 10 | 1.00 | 15.00 | −6.844 | 0.019 | 4.748 | 0.553 | 0.713 | 0.021 |
| 11 | 1.00 | 30.00 | −5.856 | −0.169 | 4.629 | 0.637 | 1.189 | 0.093 |
| 12 | 0.20 | 0.00 | −3.731 | 0.906 | 0.803 | 0.681 | 0.051 | 0.002 |
| 13 | 0.60 | 15.00 | −3.731 | 0.906 | 0.803 | 0.681 | 0.378 | 0.014 |
| 14 | 0.80 | 30.00 | −5.516 | −0.165 | 4.244 | 0.663 | 0.825 | 0.078 |
| 15 | 0.80 | 15.00 | −6.540 | 0.051 | 4.393 | 0.562 | 0.550 | 0.018 |
| 16 | 0.40 | 15.00 | −5.011 | 0.235 | 2.826 | 0.633 | 0.212 | 0.012 |

calculated viscosity at 50 s^{-1} and 10^5 s^{-1} denoted as η_L and η_H , respectively.

The calculated viscosity values were then used to generate the models for the low shear and high shear viscosity as a function of polymer concentrations with the experimental design software. The results are shown as Equations (3) and (4), respectively. Contour plots showing how the concentration of xanthan and dextran affect both low and high shear viscosity, and demonstration of the high correlation between predicted and experimental data ($R^2 = 0.97$ and $R^2 = 0.99$ for low shear and high shear viscosity, respectively) are provided in the supplementary electronic file (Fig. S1 and Fig. S2).

$$\sqrt{\eta_{50}} = 0.11 + 0.65[\text{Xan}] + 0.00093[\text{Dex}] + 0.00022[\text{Xan}]^2 + 0.0045[\text{Xan}][\text{Dex}] \quad (3)$$

$$\log[\eta_{10^5}] = -1.85 + 0.18[\text{Xan}] + 0.63[\text{Dex}] + 0.066[\text{Dex}]^2 - 0.058[\text{Xan}][\text{Dex}] \quad (4)$$

[Xan] and [Dex] indicate the concentration of xanthan and dextran, respectively.

The model for viscosity at 50 s^{-1} (Equation (3)) includes a linear and quadratic term for the concentration of xanthan and an interaction term for the two thickener concentrations. The model was highly significant ($p < 0.001$) with adjusted R^2 and predicted R^2 values of 0.99 and 0.97, respectively, and an 'adequate precision' (signal-to-noise ratio) of 48.28. These statistics indicate a robust model that describes variation across the design space well. The model for viscosity at high shear rate (Equation (4)) includes a linear term for the concentration of xanthan, a linear and a quadratic term for the concentration of dextran, and an interaction term for the two thickeners concentrations. The model was highly significant ($p < 0.001$) with adjusted R^2 and predicted R^2 values of 0.99 and 0.99, respectively, and an 'adequate precision' of 109.06.

The models reveal that the concentration of xanthan has a large effect on the low shear viscosity of the samples while the concentration of dextran has little effect. On the other hand, the concentration of dextran impacts to a larger extent on the high shear viscosity than the concentration of xanthan.

3.2. Study sample composition and shear rheological behaviour

Based on the two models, two groups of five different samples respectively with the same low shear (P-1 ... P-5, Group 1) and high shear (P-6 ... P-10, Group 2) viscosity were designed. Their measured viscosity behaviour is shown in Figs. 1 and 2. This was also fitted with the log-log model (Equation (1)) and the fitting parameters including calculated viscosity are shown in Table 3.

The results highlight the impact of dextran on high shear viscosity and of xanthan gum on the degree of shear thinning. Group 2 samples are less viscous compared to Group 1 samples except for P-8 which was most shear thinning of all samples.

3.3. Extensional flow behaviour of the designed samples

The characteristic behaviour of the ten samples in predominantly extensional flow was evaluated through filament thinning experiments. The results are shown as the evolution of the normalised filament diameter in Figs. 3 and 4. It is worth noting that the time scale of breakup for both sets of samples varies by one decade and therefore the results have been plotted on different x-axis scales. The results for the steady state extensional viscosity have been included in Table 2. For results of further analysis the

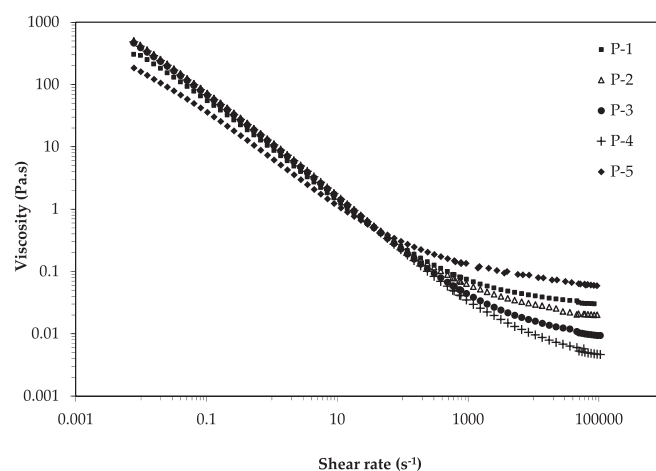


Fig. 1. Viscosity curves of low shear iso-viscous samples of Group 1.

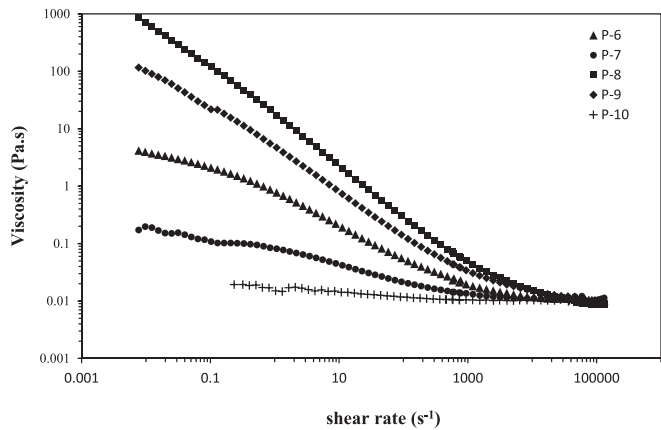


Fig. 2. Viscosity curves of high shear iso-viscous samples of Group 2.

reader is referred to the electronic supplementary material (Table S1).

The two groups of samples vary significantly in terms of breakup time. For the low shear isoviscous Group 1 samples, the breakup time ranged from 0.354 to 2.642 s while for the high shear iso-viscous Group 2 samples the breakup time ranged from 0.036 to 0.58 s. In Group 1, a sample with higher η_H equivalent to a higher concentration of dextran took longer to break up, whereas in Group 2, samples with higher η_L or a higher level of xanthan took longer to break up.

It was also found that samples with shorter breakup time, the filament thinning behaviour is characterised by fast exponential decay, e.g. P-10. Also the polymer concentrations seemed to have a large effect on the breakup time. It has been suggested that the filament thinning in the exponential decay domain was mainly due to the disentanglement and orientation of polymers (Bousfield, Keunings, Marrucci, & Denn, 1986). Therefore, in higher concentration of polymers, this domain is longer. Also it was found that the interactions between rigid rod molecular xanthan and random coil dextran could further refrain the polymer solutions from break up compared to single polymer solutions. Examples are P-4 and P-10, which had fast break up time within their groups.

3.4. Viscoelastic behaviour in small amplitude oscillatory shear

The viscoelastic moduli acquired in small amplitude oscillatory shear at increasing angular frequency (ω) are shown in Figs. 5 and 6 for low shear isoviscous samples of Group 1 and high shear

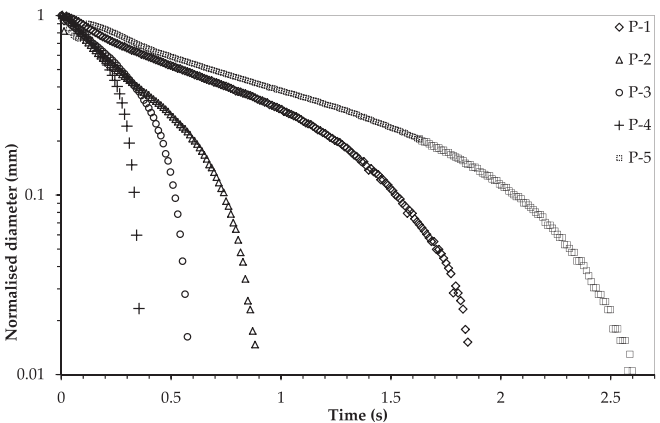


Fig. 3. Evolution of the normalised filament mid-point diameter for the low shear isoviscous Group 1 samples. Stretch time was 50 ms and measurement temperature 37 °C. Standard deviation is within 5% for all data.

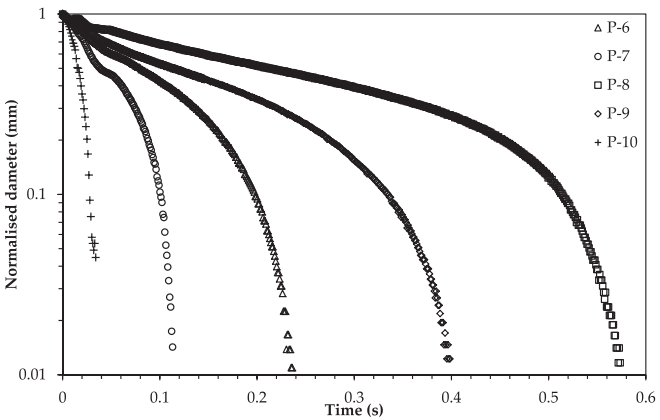


Fig. 4. Evolution of the normalised filament mid-point diameter for the high shear iso-viscous Group 2 samples. Stretch time was 50 ms and measurement temperature 37 °C. Standard deviation is within 5% for all data.

isoviscous samples of Group 2, respectively. The data were acquired at 0.1% strain which is within the linear viscoelastic domain.

Within the frequency domain analysed, all Group 1 samples showed predominantly elastic behaviour as indicated by larger G' than G'' values and can be described as weak gels. This applies only to two Group 2 samples, P-8 and P-9, which have the highest xanthan concentration out of the five Group 2 samples, within the

Table 3
Polymer concentration, log-log model parameters and calculated viscosity for the designed samples. P-1 ... P-5 form Group 1 with similar low shear viscosity (η_L) and P-6 ... P-10 form Group 2 with similar high shear viscosity (η_H). Sample 10 showed close to Newtonian behaviour and therefore the log-log model cannot be applied. The last column shows the steady state extensional shear viscosity.

| Sample no | Concentration xanthan dextran | | α | β | δ | γ | η_L | η_H | η_E |
|-----------|-------------------------------|---------|----------|---------|----------|----------|----------|----------|----------|
| | % (w/w) | % (w/w) | | | | | | | |
| P-1 | 0.61 | 22.59 | −4.907 | 0.164 | 0.722 | 3.220 | 0.374 | 0.029 | 65.165 |
| P-2 | 0.71 | 17.33 | −0.544 | 0.210 | 0.674 | 3.510 | 0.398 | 0.019 | 32.225 |
| P-3 | 0.74 | 9.40 | −6.037 | 0.311 | 0.604 | 3.610 | 0.358 | 0.009 | 14.595 |
| P-4 | 0.83 | 0.00 | −6.847 | 0.380 | 0.532 | 3.850 | 0.353 | 0.004 | 8.925 |
| P-5 | 0.40 | 32.00 | −4.348 | −0.06 | 0.731 | 3.050 | 0.42 | 0.063 | 88.143 |
| P-6 | 0.21 | 15.70 | −2.875 | 0.841 | 0.878 | 0.780 | 0.077 | 0.010 | 7.617 |
| P-7 | 0.09 | 17.11 | −1.284 | 0.950 | 0.974 | −0.730 | 0.026 | 0.010 | 9.122 |
| P-8 | 1.00 | 6.18 | −6.446 | 0.373 | 0.595 | 3.900 | 0.485 | 0.008 | 13.199 |
| P-9 | 0.47 | 12.59 | −4.965 | 0.392 | 0.669 | 2.700 | 0.214 | 0.010 | 11.482 |
| P-10 | 0.02 | 17.00 | — | — | — | — | 0.012 | 0.010 | 1.254 |

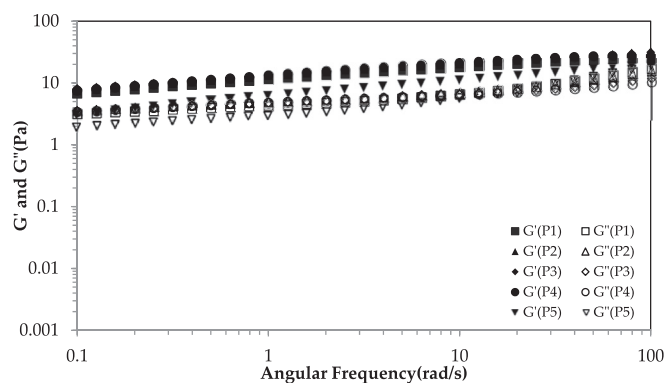


Fig. 5. Storage modulus G' (filled symbols) and loss modulus G'' (open symbols) as a function of frequency for the low shear isoviscous samples of Group 1, acquired at the strain of 0.1% and at 20 °C.

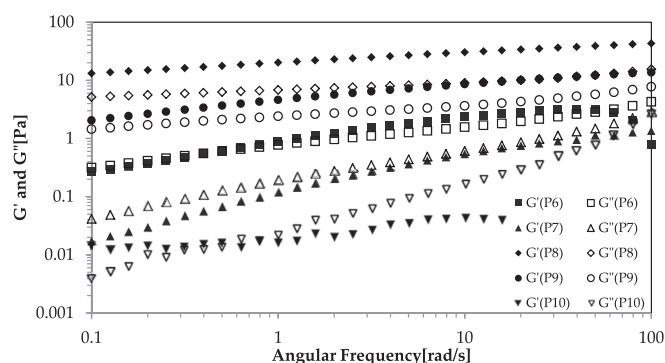


Fig. 6. Storage modulus G' (filled symbols) and loss modulus G'' (open symbols) as a function of frequency for the high shear isoviscous Group 2 samples, acquired at the strain of 0.1% and at 20 °C.

range of xanthan covered by the Group 1 samples. With decreasing xanthan concentration, the viscous material response increases. The presence of dextran has little impact on the small deformation viscoelastic behaviour which is contrary to the behaviour in extensional flow.

The complex viscosity as a function of angular frequency is shown in Figs. 7 and 8 for the low shear isoviscous Group 1 and the high shear isoviscous Group 2 samples, respectively. Complex viscosity curves were similar for Group 1 samples which was expected since they had similar viscoelastic moduli. Only sample P-5 showed a slightly lower than other samples in Group 1 due to the lower concentration of xanthan. Also, the complex viscosity of Group 1

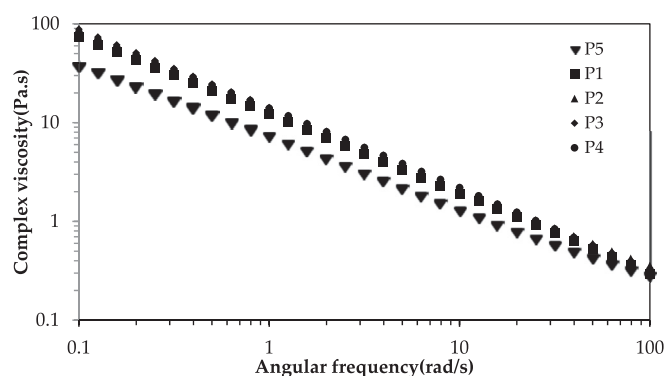


Fig. 7. Complex viscosity as a function of angular frequency for Group 1 samples.

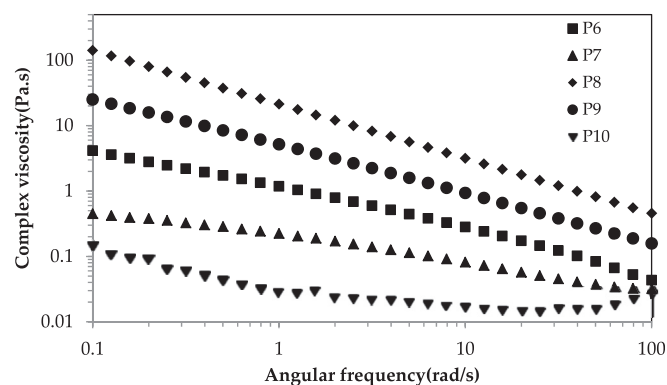


Fig. 8. Complex viscosity as a function of angular frequency for Group 2 samples.

samples showed a very steep frequency dependence, which again is indicative of weak gel behaviour. As a comparison, complex viscosity of Group 2 samples showed large differences due to the differences in xanthan concentration. With reduced xanthan concentration, complex viscosity as well as dependence on angular frequency were reduced.

4. Sensory properties and correlation with sample rheology

4.1. Panel performance

Table 4 shows the results of the mean scores of the sensory tests which are discussed in the following sections. To judge panel performance, one needs to analyse the P -value also shown in Table 4. It can be seen that both products and panellists were significantly different ($p < 0.05$). The results show that there were some interactions between samples and panellists for all of the mouthfeel attributes. However, when checking the product*panellists interaction plot (results not shown), it was found that these interactions, although statistically significant, were of no real consequences. This was simply due to the slight differences in the use of the scale by some panellists. For the overall fruity flavour and overall sweetness, there were some crossover effects discussed below.

4.2. Mouthfeel perception and correlation with shear and extension viscosity

The ANOVA results show that for samples with either similar low shear or high shear viscosity, the perceived mouthfeel perceptions were significantly different. If only low shear or high viscosity were used in predicting mouthfeel perceptions, the results would not be accurate because even if the samples have the same low or high shear viscosity, they will still be perceived as different in terms of mouthfeel perception. This is clear indication for the importance of considering both low and high shear viscosity when correlating shear viscosity with mouthfeel perceptions.

'Initial Thickness in Mouth' were highly correlated ($r = 0.998$). Mean sample scores (Table 4) show that within each group (Group 1 and Group 2), all five samples were perceived as significantly different to each other for the 'Thickness' attributes, despite being identical in either low or high shear viscosity. When performing a correlation between the physical measurements of viscosity with the sensory score, perceived initial thickness was highly correlated to viscosity at low shear rate ($r = 0.961$) but less well correlated to high shear viscosity ($r = 0.556$) (see Table 5). Similarly, 'Thickness in mouth' had a correlation coefficient of 0.952 and 0.577 with low and high shear viscosity, respectively. While the correlation coefficient with low shear viscosity was higher than for

Table 4
ANOVA associated p-values and mean sample scores for sensory attributes for samples. P-1 ... P-5 form Group 1 with similar low shear viscosity (η_L) and P-6 ... P-10 form Group 2 with similar high shear viscosity (η_H).

| | | Initial thickness | Thickness in mouth | Stickiness on lips | Stickiness in mouth | Mouth coating | Overall fruity flavour | Overall sweetness |
|-------------------|-------------------|--------------------|---------------------|---------------------|---------------------|-------------------|------------------------|-----------------------|
| p-values | Samples | <0.001 | <0.001 | <0.001 | <0.001 | <0.001 | 0.002 | <0.001 |
| | Panelists | <0.001 | <0.001 | <0.001 | <0.001 | <0.001 | <0.001 | <0.001 |
| | Samples*Panelists | <0.001 | <0.001 | <0.001 | 0.0052 | 0.0012 | 0.0197 | 0.275 |
| Mean sample score | P-1 | 8.67 ^{b*} | 8.32 ^b | 8.06 ^a | 7.77 ^b | 7.63 ^b | 5.45 ^{a,b} | 6.5 ^{b,c} |
| | P-2 | 7.68 ^c | 7.41 ^c | 6.56 ^{b,c} | 6.41 ^c | 6.56 ^c | 5.84 ^{a,b} | 6.34 ^{b,c,d} |
| | P-3 | 6.49 ^d | 5.99 ^d | 5.91 ^c | 5.06 ^d | 4.86 ^d | 4.86 ^{b,c} | 5.02 ^{d,e} |
| | P-4 | 5.29 ^e | 4.60 ^e | 3.91 ^d | 3.14 ^e | 3.66 ^e | 3.23 ^d | 2.94 ^f |
| | P-5 | 9.26 ^a | 9.05 ^a | 8.85 ^a | 8.79 ^a | 8.52 ^a | 6.34 ^{a,b} | 7.60 ^{a,b} |
| | P-6 | 1.83 ^g | 1.44 ^f | 2.66 ^e | 1.70 ^f | 1.56 ^f | 6.13 ^{a,b} | 6.99 ^{a,b,c} |
| | P-7 | 0.92 ^h | 0.84 ^g | 2.08 ^{e,f} | 1.41 ^f | 0.99 ^g | 6.62 ^a | 7.34 ^{a,b} |
| | P-8 | 8.09 ^c | 7.85 ^{b,c} | 6.88 ^b | 6.65 ^c | 6.52 ^c | 3.8 ^{c,d} | 3.78 ^{e,f} |
| | P-9 | 4.37 ^f | 4.09 ^e | 3.97 ^d | 3.60 ^e | 3.30 ^e | 5.40 ^{a,b} | 5.78 ^{c,d} |
| | P-10 | 0.54 ^h | 0.42 ^g | 1.52 ^f | 0.65 ^g | 0.64 ^g | 6.87 ^a | 8.01 ^a |

* Sample coded with the same letter in any one column are not significantly different with respect to this attribute ($p > 0.05$).

Table 5
Correlation between viscosity at low and high shear rate, extensional viscosity (η_e) and mouthfeel sensory scores.

| | Correlation coefficient R ² | | | | |
|---|--|--------------------|--------------------|---------------------|---------------|
| | Initial thickness | Thickness in mouth | Stickiness on lips | Stickiness in mouth | Mouth-coating |
| η at 50 s ⁻¹ | 0.961 | 0.952 | 0.890 | 0.884 | 0.911 |
| η at 10 ⁵ s ⁻¹ | 0.556 | 0.577 | 0.670 | 0.688 | 0.663 |
| η_e | 0.862 | 0.872 | 0.902 | 0.909 | 0.911 |

high shear viscosity, which is in agreement with Wood (1968) who reported that viscosity at 50 s⁻¹ relates to thickness perception, generating models with or without inclusion of high shear viscosity led to higher correlation coefficients for the latter. The models and correlation coefficients are reported in Table 6.

As can be seen from Table 6, the models built for predicting thickness perceptions without η_H featured linear relationships with η_L with R² of 0.92 and 0.91 for 'Initial thickness' and 'Thickness in mouth', respectively. The predicted R² for 'Initial thickness' and 'Thickness in mouth' was 0.889 and 0.867, respectively. The adjusted R² were 0.913 and 0.894 for 'Initial thickness' and 'Thickness in mouth' respectively. The Adequate precision was 19.686 and 17.581 for 'Initial thickness' and 'Thickness in mouth', respectively. In order to further illustrate the model, the experimental results are plotted against values that have been predicted from the models and the results are shown in Fig. 9(a and c). As can be seen from the figure, the predicted and experimental values are perfectly matched for samples with sensory scores approximately below 5. This indicates that for samples that were perceived as less thick, it is probably the low shear viscosity that mainly decided the 'thickness' perceptions. However, it seems that as the score for perceived thickness increased above 5, there are some deviations between the predicted and experimental values. This indicates that perhaps for samples that are perceived as more 'thick', viscosity at low shear rate is not solely sufficient in predicting the sensory scores.

As a comparison, models that included both low and high shear viscosity were also examined. As can be seen from Table 6, models

including both low and high shear viscosity featured linear relationships with both low and high shear viscosity and also a quadratic relationship with high shear viscosity. All the model description parameters were largely increased which indicates that the models are more robust compared to models that only including low shear viscosity. A further illustration of the models can be found in Fig. 9(b and c). Correlation to Kokini oral shear was not considered in this study since calculation of the Kokini oral shear stress requires the viscosity behaviour to be described by the power law model and this is not the case for the xanthan and dextran based samples.

The attributes 'Stickiness on the lips' and 'Stickiness in mouth' were highly correlated r ($r = 0.997$). The results also show that within Group 1 and Group 2, all 5 samples were perceived as significantly different in terms of 'Stickiness on lips' and 'Stickiness in mouth'. Correlation of the sensory scores to viscosity at low shear and high shear over all 10 samples revealed that the sensory scores were better correlated to low shear viscosity ('Stickiness on the lips' $r = 0.890$ and 'Stickiness in mouth' $r = 0.884$) but less well correlated to high shear viscosity ('Stickiness on the lips' $r = 0.670$ and 'Stickiness in mouth' $r = 0.688$). It is worth pointing out that for the perception of 'Stickiness', the correlation coefficient between high shear viscosity and sensory score is higher than for the perception of 'Thickness'. Although the level of correlation is poor, it may be worth pointing out that the better correlation of 'Stickiness' to high shear viscosity implies the possibility of high shear rates playing a more important role in the process of evaluating 'Stickiness' than

Table 6
Comparisons of prediction models for thickness perception with and without viscosity at high shear rate.

| | | R ² | Adj. R ² | Pred. R ² | Adeq. precision |
|-------------------------|--|----------------|---------------------|----------------------|-----------------|
| Models without η_H | Initial thick = $0.44 + 17.95 \cdot \eta_L$ | 0.920 | 0.913 | 0.889 | 19.686 |
| | Thick in mouth = $0.23 + 17.55 \cdot \eta_L$ | 0.910 | 0.894 | 0.867 | 17.581 |
| Models with η_H | Initial thick = $-0.98 + 16.3 \cdot \eta_L + 170 \cdot \eta_H - 1839.4 \cdot \eta_H^2$ | 0.996 | 0.993 | 0.936 | 50.320 |
| | Thick in mouth = $-1.3 + 15.8 \cdot \eta_L + 185 \cdot \eta_H - 1993 \cdot \eta_H^2$ | 0.995 | 0.992 | 0.971 | 47.700 |

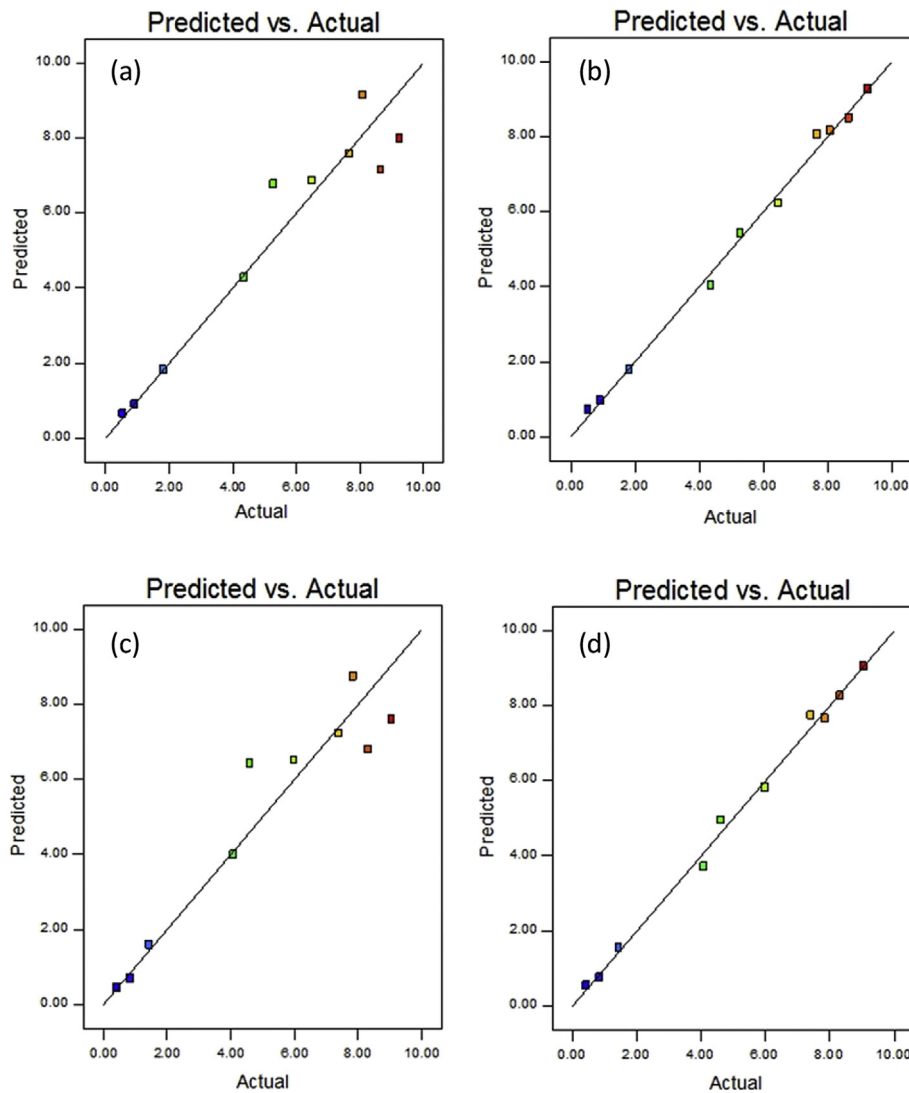


Fig. 9. Comparisons of predicted values from models that with/without η_H and experiment values: (a) Initial thickness without η_H ; (b) Initial thickness with η_H ; (c) Thickness without η_H ; (d) Thickness with η_H .

'Thickness' of thickener solutions. These higher shear rates would be a result of the attribute evaluation protocol, this is to some extent surprising as it has been postulated that shear rates in the narrow gap between tongue and palate can reach very high values. Considering the evaluation protocols of 'Stickiness', it seems obvious to inspect the relationship between the sensory scores for 'Stickiness' and extensional viscosity as determined by filament break-up. Indeed, the extensional viscosity was found to be well correlated to 'Stickiness' ($r = 0.902$ and $r = 0.909$ for 'Stickiness on the lips' and 'Stickiness in mouth', respectively), better even than the low shear viscosity, as can be seen from Table 5.

When building models to predict the sensory perceptions of 'Stickiness' and 'Mouth coating', models that only included low shear viscosity and extensional viscosity predicted the perception better than models including all three factors or only low and high shear viscosity (see Equations in Table 7). While 'Stickiness' was highly correlated to 'Thickness' ($r = 0.983$), and similar results have been reported previously (Morris et al., 1984), including extensional viscosity in the models for 'Thickness' did not better the predictive power.

There are few reported studies that have employed CaBER or

extensional viscosity measurements to study the stickiness perception of foods. Similar results can be found in Chen, Feng, Gonzalez, and Pughaloni (2008) who studied the relationship between the tensile force of foods and their sensory scores for stickiness evaluated by a 'finger separation' experiment. The authors suggested that the maximum tensile force and the work till the maximum force were two useful parameters for predicting food stickiness. The results of this research based on in-mouth assessment of 'Stickiness' and a rheological assessment in filament break-up confirm the suggestions made by Chen et al. (2008), including a correlation equation with increased predictive power when including extensional.

'Mouth coating' followed the same trend as 'Stickiness'. Scores given to samples within each of the two sample groups differed and the model with the highest correlation coefficient for predicting this sensory attribute also involved both low shear viscosity and extensional viscosity. This result is another indicator that the flow situation in the oral cavity is highly complex and should be described as a superposition of shear and extensional flow.

Table 7
Prediction models for Stickiness and Mouth coating and model descriptors.

| | R ² | Adj. R ² | Pred. R ² | Adeq. precision |
|--|----------------|---------------------|----------------------|-----------------|
| Stickiness on lips = $1.42 + 9.26\eta_L + 0.04\eta_E$ | 0.965 | 0.955 | 0.928 | 25.460 |
| Stickiness in mouth = $0.54 + 10.1\eta_L + 0.05\eta_E$ | 0.963 | 0.952 | 0.923 | 24.640 |
| Mouth coating = $0.34 + 10.82\eta_L + 0.05\eta_E$ | 0.981 | 0.975 | 0.955 | 34.293 |

4.3. Mouthfeel perception and correlation with small amplitude oscillatory shear properties

Correlations of complex viscosity values taken at different angular frequency to the sensory attributes evaluated in this study are shown in Table 8. In case of the attributes of 'Initial thickness' and 'Thickness in mouth' the sensory scores were better correlated to complex viscosity at higher angular frequency ($r = 0.89$ at 100 rad/s) than with the values taken at lower angular frequency, this observation is in agreement with literature (Richardson et al., 1989).

As can be seen from Table 8, with increased frequency the correlation between complex viscosity and mouthfeel perception increased and reached the highest value for all attributes at frequency of 100 rad/s. For 'Thickness' perceptions, the correlation between sensory scores and complex viscosity at 100 rad/s were highest ($r = 0.89$) among all the attributes, followed by 'Mouth coating' ($r = 0.83$) and 'Stickiness' ($r = 0.81$ and $r = 0.8$ for 'Stickiness on lips' and 'Stickiness in mouth', respectively). The results are in accordance with Richardson et al. (1989) who found that mouthfeel perceptions were best correlated to complex viscosity at 50 rad/s. They also suggested that for 'weak gels' such as xanthan solutions, oral evaluation was predominantly based on viscoelastic properties of the intact network structure rather than on those of the isolated species released after rupture of the network by shear.

Interestingly it was found that for the high shear isoviscous samples of Group 2, complex viscosity was highly correlated to mouthfeel perceptions at all frequencies ($r > 0.95$) with the highest correlation coefficient $r = 0.98$ for all attributes occurring at frequency of 50 rad/s. However, the correlation between complex viscosity and mouthfeel attributes for the low shear isoviscous samples of Group 1 were relatively poor and it seems that samples with higher complex viscosity were perceived as lower in terms of mouthfeel perceptions. These results may indicate that complex viscosity is a useful predictor for mouthfeel perceptions for samples that behave significantly different under small deformations. These samples covered the range from true solutions to samples that show 'weak gel' properties. However, the results from this research clearly indicate that for samples that behave similarly under small deformation, complex viscosity cannot be used to predict the mouthfeel, and properties under large deformation maybe more relevant to their mouthfeel perception.

Table 8
The correlation between complex viscosity at different angular frequency and sensory scores of mouthfeel attributes.

| | Correlation coefficient (r) | | | | |
|-----------------------|-----------------------------|--------------------|--------------------|---------------------|---------------|
| | Initial thickness | Thickness in mouth | Stickiness on lips | Stickiness in mouth | Mouth-coating |
| η^* at 0.1 rad/s | 0.74 | 0.73 | 0.63 | 0.62 | 0.66 |
| η^* at 1 rad/s | 0.78 | 0.76 | 0.66 | 0.65 | 0.69 |
| η^* at 10 rad/s | 0.81 | 0.79 | 0.69 | 0.68 | 0.72 |
| η^* at 50 rad/s | 0.86 | 0.85 | 0.76 | 0.75 | 0.78 |
| η^* at 100 rad/s | 0.89 | 0.89 | 0.81 | 0.80 | 0.83 |

4.4. Flavour and sweetness perception

In the evaluation of flavour and taste, some interesting results were found and are reported here. It should be noted that the c/c^* ratio was not considered in the data analysis as the samples corresponded to mixtures of polymers rendering this ratio somewhat meaningless. The average maximum intensity (Imax) and cumulative area (AUC) for the ion monitoring IAA (ion 131) for the 10 samples are shown in Fig. 10. It was found that for both Imax and AUC, there were no significant differences across the samples ($p > 0.05$). However, as part of the sensory evaluation of flavour, the scores for the overall fruity flavour (see Table 4) tell a different story as discussed in the following subsequent to the scores for overall sweetness.

It can be seen from the sensory results reported in Table 4 that, despite of all the samples having the same level of sucrose (3%), the overall scores for 'Sweetness' ranged from 2.91 to 8.01. For the low shear isoviscous samples of Group 1, it was found that higher scores of 'Overall sweetness' were given to samples that scored higher in terms of mouthfeel perceptions. It is generally believed that perceived taste is decreased with increased viscosity (Baines & Morris, 1987; Malkki et al., 1993; Christensen, 1980; Cook, Hollowood, Linforth, & Taylor, 2003) and also that different hydrocolloids affect sweetness to different extents (Pangborn & Szczesniak, 1974; Vaisey, Brunon, & Cooper, 1969). The results from this research seem to somewhat disagree with the results from these previous studies. However, it is worth noting that for

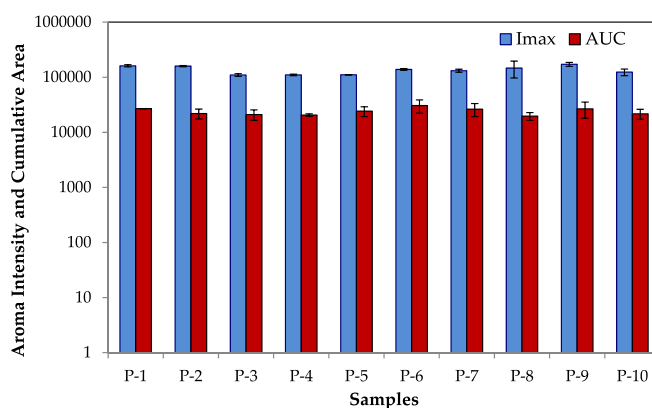


Fig. 10. Maximum aroma intensity (Imax) and Cumulative area (AUC) of the *in-vivo* flavour release.

samples in Group 1, samples that were higher in sweetness perception contained lower concentrations of xanthan and higher concentration of dextran. This rule also seemed true across the whole samples set. For samples with the same levels of xanthan, such as P5 (0.4%) and P9 (0.47%), the one with higher dextran was given higher scores of 'Sweetness'. The results indicated that within the design space, the concentration of dextran and xanthan have opposing effects on the perception of sweetness.

The results for Group 1 samples also indicate that the perceived sweetness may be affected to a lesser extent in samples that are less shear thinning. The relationship between rheological behaviour of hydrocolloids and their sweetness perceptions were also reported by Vaisey et al. (1969), and they found that hydrocolloid solutions that were more shear thinning tend to mask sweetness to a smaller extent. However, this research only compared the time needed for different hydrocolloid solutions to be perceived as sweetness, but not the overall intensity of sweetness.

As discussed previously, the addition of dextran will increase the high shear viscosity, elasticity and extensional viscosity of samples. Therefore, at either similar xanthan concentration or similar low shear viscosity, the samples with increased elasticity or extensional viscosity were perceived as sweeter. Elasticity and saltiness perception has been studied using Boger fluids by Koliandris et al. (2011) but found no significant difference in terms of saltiness and mouthfeel perception between Boger fluids and inelastic viscous reference samples. However, as Boger fluids are almost shear independent materials (James, 2009), it is very difficult to say how the elasticity affects the taste and mouthfeel perceptions for shear thinning materials.

Overall flavour perception was found to be highly correlated to sweetness perception ($r = 0.98$). This indicates that these two perceptions interacted with each other. Results from APCI-MS as seen in Fig. 10 indicate that during consumption of the samples, both the maximum intensity of flavour released and the total amount of flavour released were not significantly different between samples ($p > 0.05$). This revealed that it was the perception of sweetness that affected the perception of flavour. Indeed, the interactions between volatile and non-volatile stimuli are well documented (Davidson, Linforth, Hollowood, & Taylor, 1999; Hewson, Hollowood, Chandra, & Hort, 2008; Hollowood et al., 2002; Hort & Hollowood, 2004; Pfeiffer, Hort, Hollowood, & Taylor, 2006; Taylor, Hollowood, Davidson, Cook, & Linforth, 2002). Davidson et al. (1999) found that the reduction of perceived mint flavour was correlated to decreased sugar release in chewing despite the fact that release of mint volatile remained constant. Hollowood et al. (2002) suggested that the perception of flavour was reduced not because of the reduced flavour release but due to the reduced sweetness perception, and they hypothesised that it was due to the increased concentration of hydrocolloid that reduced the amount of free water to carry tastants to the receptors.

5. Conclusions

This research has validated that mouthfeel, aroma and taste perceptions of hydrocolloid thickened solutions are highly correlated to viscosity at low shear rate (50 s^{-1}). However, analysing the power of predictive models has shown that it can be enhanced by considering further rheological parameters. Thickness perception was found to be better predicted through a model including both low and high shear viscosity. For stickiness and mouth coating perceptions, better predictions could be made through models including both low shear and extensional rheology. It was also found that mouthfeel perceptions were highly correlated to the complex viscosity measured at 100 rad/s . *In-vivo* flavour release indicated that there were no significant differences between

maximum flavour intensity and total flavour release. However, sensory scores for overall sweetness and flavour were highly correlated to each other. Since sweetness perception is strongly affected by the viscosity of a sample, therefore it is the interaction between viscosity, sweetness and flavour that affect the final perception of samples.

In conclusion, there is no single rheological parameter that will ultimately correlate to a range of mouthfeel perceptions. Depending on the mouthfeel perception or rather the evaluation protocol applied during the scoring of a particular mouthfeel parameter, a model comprising shear and extensional rheological parameters will have higher predictive power than a model solely based on shear rheological parameters. It is worth noting though that this conclusion is based on working with a thickener system containing xanthan, which possesses a rod-like molecular structure giving it its highly shear thinning property but also making it unique among food thickeners potentially limiting transferability of the models developed here to other thickener systems.

Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.foodhyd.2016.05.010>.

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